

# Recycling of Natural Fibre Thermoplastic Compounds

*A. Ramzy, A. Elsabbagh, L. Steuernagel, D. Meiners, G. Ziegmann*

*Institut für Polymerwerkstoffe und Kunststofftechnik / TU Clausthal*

*Amna.ramzy@tu-clausthal.de*

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## Abstract

Plastics and polymer industry have gained a bad reputation regarding the environmental issues as being responsible of several problems especially climate, energy resources and landfills. That is why focus is switched to natural fibres as an alternative to synthetic fibres. This research studies the influence of recycling on injected moulded thermoplastic natural fibre reinforced parts regarding the thermal, thermo-mechanical behaviour and change in fibre content and shape. Two polypropylene compounds are investigated namely PP – sisal and PP – hemp compounds, where the two polypropylene matrices are modified with MAPP. Processing cycles proved to decrease the fibre content of both compounds where PP-sisal (PP-S) suffers a reduction of fibre content by 26.5% whereas the PP-hemp (PP-H) compound decreased by only 10.2% of the initial value. An increase of 7.2°C and 11.6°C in the T<sub>g</sub> (Glass transition temperature) is recorded for the compound PP-S and PP-H respectively. This research has proved the high potential in recycling natural fibres compound since they show improvement in flow and thermal properties after recycling for at least 5 cycles.

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## 1 Introduction

Plastics and polymer industry has gained a bad reputation regarding the environmental issues as being responsible of vital problems such as climate, energy resources and landfills. As an effective and practical approach to reduce the negative prints of polymers compounds, the introduction of natural fibres as reinforcement in engineering and industrial products is considered as an environmental friendly option. Natural fibres are well known for their technical applications over centuries in various forms from complicated structures like ships and boats and some war instruments like catapults to simple constructions like arcs and finally easy applications like flags and sails [1, 2, 3]. The recycling of materials used in industrial applications / products is a very vital topic nowadays [4]. The urgency of many current climate and surrounding catas-

trophes led to the growing attention towards recycling. Some of the environmental crises include the climate changes due to increased emission of CO<sub>2</sub>, the Ozone hole and the continuous demolishing of the rainforests. In addition, the limitation of oil supplies caused the increase in the usage of atomic power plants which in turn produce harmful waste products. This research studies the influence of recycling on injected moulded thermoplastic natural fibre reinforced plastics regarding the flow behavior, thermal and thermo-mechanical behaviour in addition to change in fibre content and shape. Two compounds are investigated namely PP – S and PP – H compounds, where the two polypropylene matrices are modified with MAPP.

## 2 Experimental

### 2.1 Reinforcing fibres and matrices:

The reinforcing fibres and matrix properties of the two natural fibre thermoplastic compounds used in this research are stated in Tab.1.

*Tab.1: Matrices and reinforcing fibres properties[1]*

Material	Melt Flow Rate (MFR) at 230°C / 2.16 Kg [g/10 min]	Tensile Modulus [MPa]	Tensile stress at yield [MPa]	Impact at 0°C [kJ/m <sup>2</sup> ]
Moplen EP 3307®	15	1100	20	10
Moplen EP 500V®	100	1450	26	5
Hemp		800	65	
Sisal		12	600	

### 2.2 Injection and recycling

Samples are prepared using the normal injection process and recycled over five cycles. Recycling is performed by shredding the injected samples then drying in oven for 24 hours at 105°C then reinjected again to start a new cycle.

The injection machine used in samples preparation is Arburg Allround 220S 150-60 where a larger injection machine Arburg Allround 320C 600-250 is used to inject the flow length spirals. All samples are injected at 500 bar, 20 ccm and 200°C.

### 2.3 Flow length

To investigate the flow behavior of the recycled compounds, a spiral mould is used in which the molten compound is injected and the flow length of the material is measured using a special template after cooling as shown in Fig. 1.

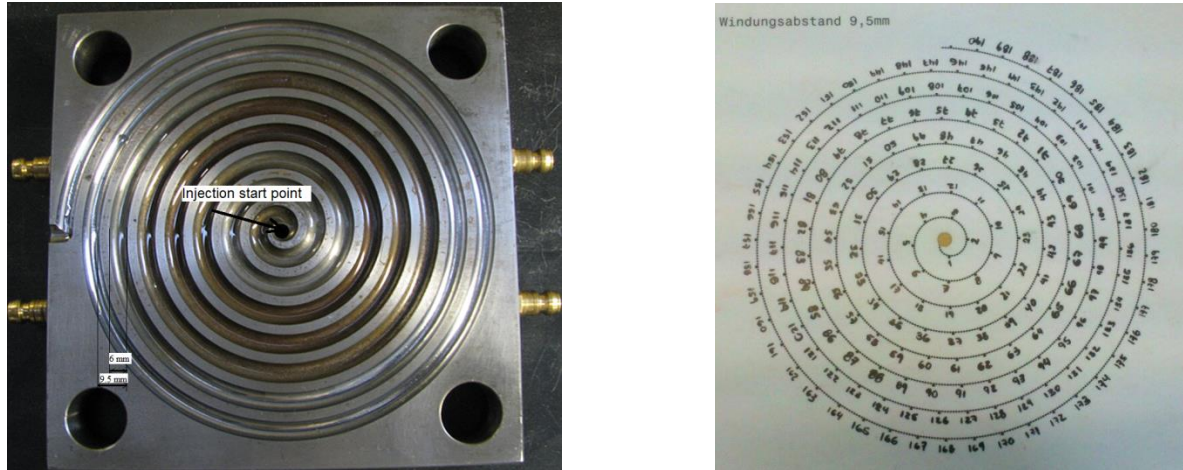


Fig. 1: Spiral mould and flow length measuring template [5]

### 2.4 Characterization

#### 2.4.1 DMA test and calculating the activation energy at $T_g$

Samples are investigated for the change in thermo-mechanical properties after recycling. This is performed using the dynamic mechanical analysis test (DMA). For this test the samples are subjected to a changing mechanical loading at defined frequencies: 0.5, 1, 2.5 and 10 Hz. Over the testing period a temperature ramp is applied from  $-100^{\circ}\text{C}$  to  $150^{\circ}\text{C}$  at constant heating rate of  $2^{\circ}/\text{min}$ . Change in storage and loss modulus are recorded to obtain the final graph.

The activation energy required for the material to undergo interphase transitions is also calculated in relation to the change in loss modulus according to Arrhenius Equation (1) as follows:

$$\ln(f) = E_a / RT \quad (1)$$

Where:

$E_a$ : Activation energy

$R$ : Gas constant

$T$ : Temperature

$f$ : Frequency

### 2.4.2 Fibre content

Fibres are extracted by means of a matrix solvent namely decaline, to investigate the effect of recycling on fibre content (FC) and length distribution. Extracted fibres are then dried and weighed in comparison with the weight before extraction as in equation (2). The change in weight gives an indication about the degree of fibre damage and degradation as a result of recycling.

$$FC\% = \frac{W_b - W_a}{W_b} \cdot 100 \quad (2)$$

Where:

$W_a$ : weight before

$W_b$ : weight after

## 3 Results and Analysis

### 3.1 Flow length

From the experiments it is proved that flow properties of the compounds are positively enhanced by the successive shredding and injection processes which is easily distinguished by the increase in the spiral flow length as shown in Fig. 2.

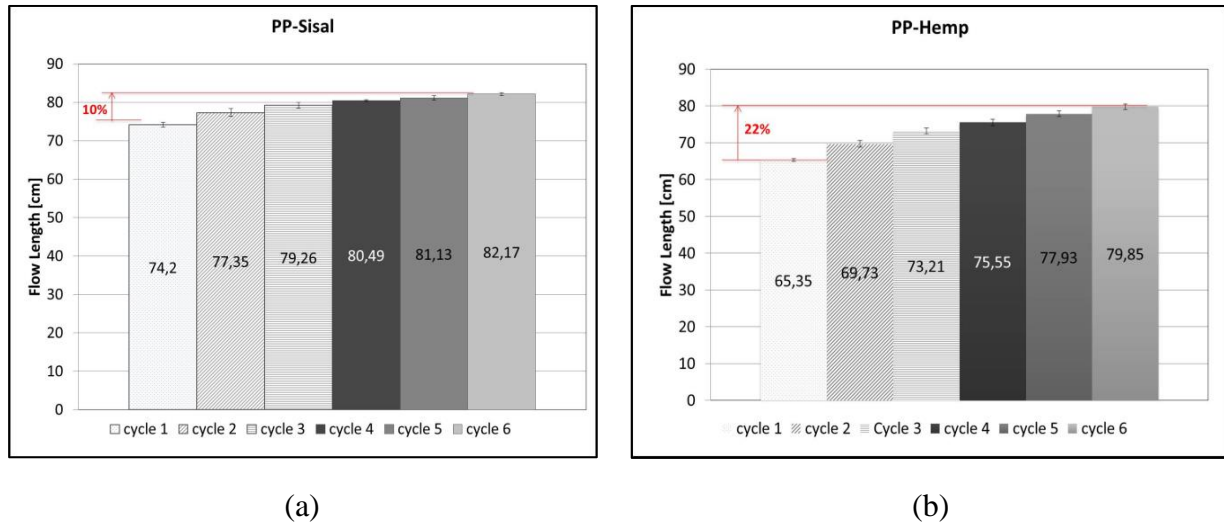


Fig. 2: Evolution of flow length for composites after recycling a- PP-Sisal b- PP-Hemp

From the graph it can be noticed that the flow length for the high flow compound (PP-Sisal) shows less sensibility to recycling in comparison to the high impact compound (PP-Hemp). This is easily distinguished from the percentage of increase in the flow length where only 10% increase in the PP-Sisal is recorded compared to over 20% increase in the high impact compound.

This could be attributed to several aspects, firstly the change in the matrix properties as a result of the severe successive processing conditions such as shredding and reinjection. Especially reinjection and the high shearing stresses applied on the material between the double screws of the injection machine under high temperatures is believed to cause chain scissions which reflect on better flow properties. For the high impact compound, it is a high crystalline compound which therefore contains by nature longer chains as by the high flow which contains shorter chains. Therefore the chain scission shows more effect on the PP-Hemp as by the PP-Sisal which already has shorter chains. The other effect comes from fibres damage, shortening and splitting.

### 3.2 DMA and Activation energy

The graph of the DMA test is analysed and Tg peaks are defined at different frequencies. Natural logarithm of the frequencies is plotted against the temperature reciprocal as shown in Fig. 3.

The activation energy is then calculated using the slope of the fitting line. As seen in Figure 2, the activation energy (trend line slope) decreased by recycling 4 times about 5%, whereas the reciprocal of Tg shifts to the left after recycling four times. In other words; Tg increases by recycling.

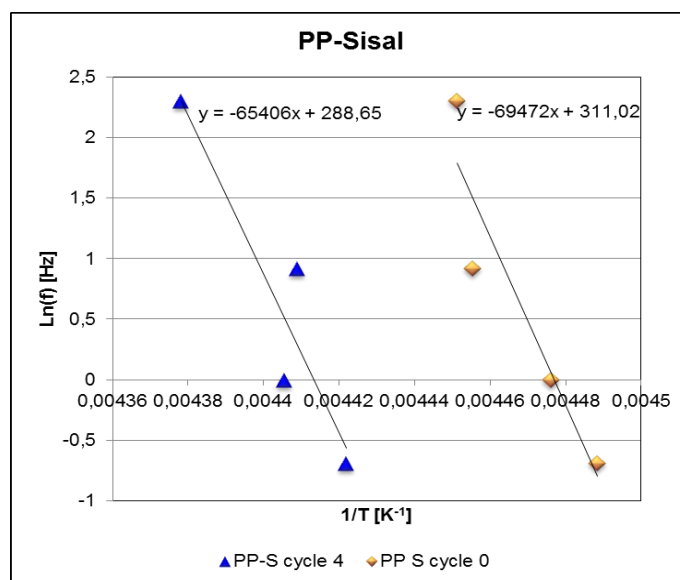


Fig. 3: Calculation of activation energy at Tg

### 3.3 Fibre content

Figure 4 shows the decrease of fibre content along the increase of recycling. Fibre content of PP-sisal is reduced by 26.5% whereas the PP-Hemp compound decreased by only 10.2%. The behaviour seems to follow a linear trend line. This decrease in the extracted fibre content is attributed to the thermal degradation of carbohydrates found in the fibres and their conversion into evolving gases as ensured by the thermal gravimetry / mass spectrometry (TG-MS) starting at 160°C as shown in Figure 5.

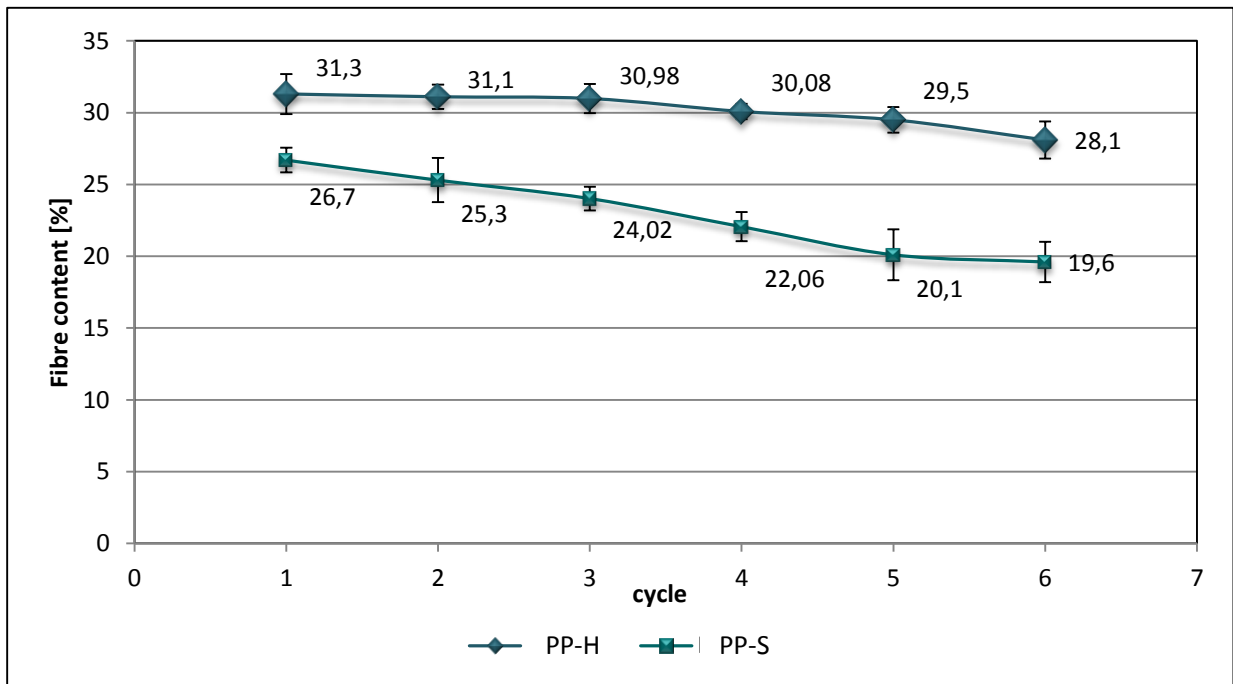


Figure 4: Fibre content of both composites at different cycles

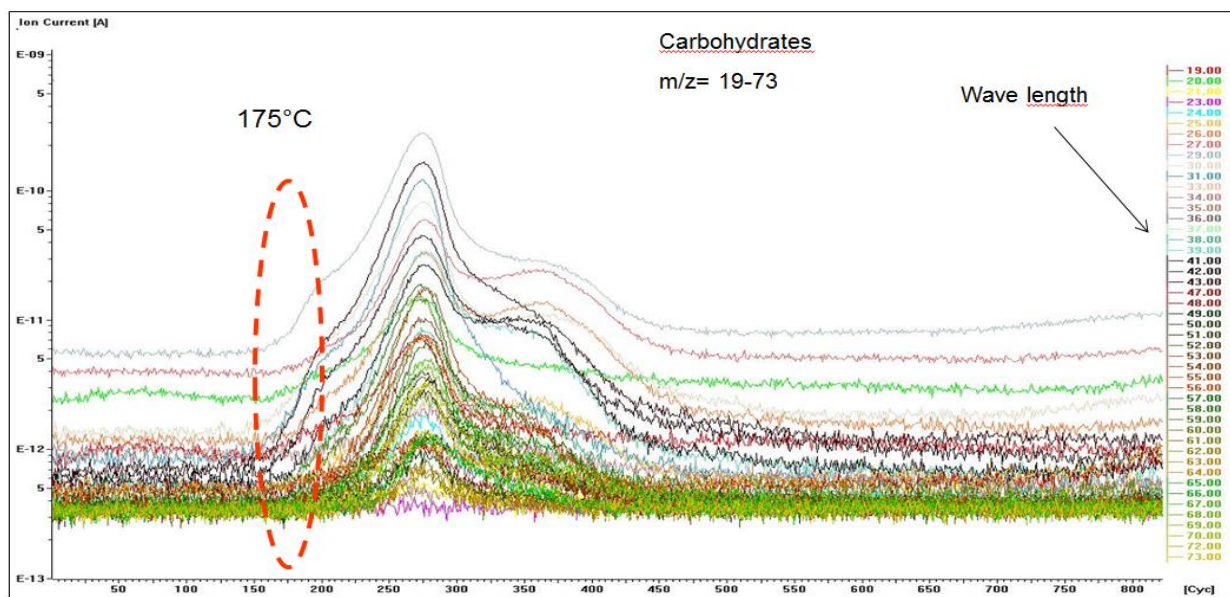


Fig. 5: Thermal gravimetry curve of carbohydrates present in PP-S and PP-H[6]



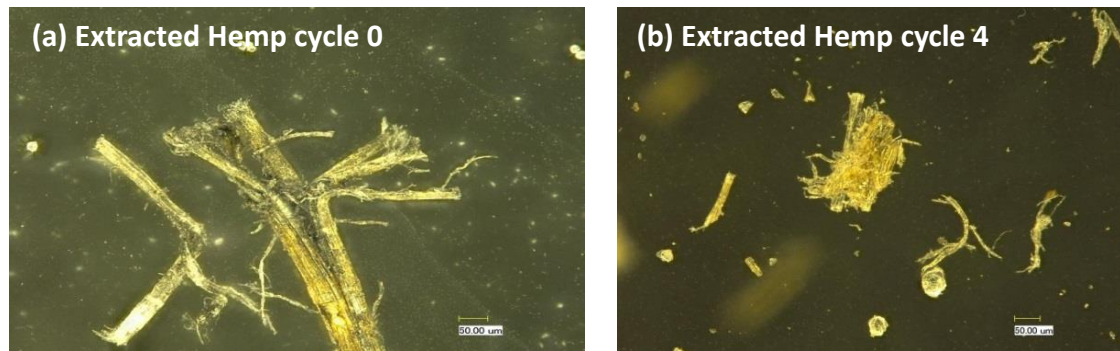


Figure 6: Fibre decomposition at (a) cycle 1 (left) (b) cycle 4 (right)

Microscopic investigation ensured the results by the fibre decomposition features as shown in Figure 6 at both the first and the fourth cycle.

#### 4 Summary

- Recycling has a positive influence on increasing the flowability of both natural fibre composites with 10% and 22% for PP-S and PP-H, respectively.
- Natural fibres decompose thermally by successive recycling. Thus the fibre content of both compounds where PP-S suffers a reduction of fibre content by 26.5% whereas the PP-H compound decreased by only 10.2%.
- Recycling results in higher Tg of 7.2°C and 11.6°C for the compound PP-S and PP-H, respectively.
- The study proved the high potential in recycling natural fibres compound as they show improvement in flow and thermal properties after recycling for at least five cycles.

#### Literature

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